

# Recent Advances in Anode Optimization for Water-Based Zinc-Ion Batteries

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**Abstract:** *With the transformation of the global energy structure to a clean and low-carbon direction, efficient and safe energy storage technology has become the focus of research. Water-based zinc-ion batteries (AZIBs) are regarded as ideal candidates for the next generation of large-scale energy storage systems due to their high safety, low cost, environmental friendliness, excellent rate performance, and cycle stability. However, the zinc cathode faces challenges such as branch crystal growth, hydrogenation reaction, and severe Corrosion during battery operation. These problems lead to poor reversibility, shortened cycle life, and low Coulomb efficiency, thereby limiting its application in energy storage and flexible electronics. This paper systematically sorts out the key problems faced by the zinc cathode recently and the corresponding optimization strategies, including the impact of interface engineering, structural design, and electrolyte regulation on the stability of the cathode, and looks forward to the future development trend of water-based zinc-ion batteries to provide guidance and inspiration for its efficient application.*

**Keywords:** Zinc-ion batteries; Anode modification; Interfacial engineering; Structural design; Electrolyte.

## 1. INTRODUCTION

Among electrochemical energy storage systems, aqueous zinc-ion batteries (AZIBs) typically feature a zinc metal anode and an aqueous electrolyte. Owing to their high power density, low cost, and environmental friendliness, AZIBs have found extensive applications in large-scale energy storage systems, portable electronics, and electric vehicles. As a type of secondary zinc-based battery, AZIBs mainly consist of a cathode, anode, electrolyte, and separator, storing and converting energy through the shuttling of  $\text{Zn}^{2+}$  ions between the electrodes.

At present, the research on AZIBs (zinc ion batteries) mainly focuses on the development, energy storage mechanism, and characterization technology of high-performance cathode materials, while the research on cathode materials is still relatively limited. However, the zinc cathode faces key challenges, such as branch crystal growth, hydrogenation reactions, and Corrosion, in actual operation. These problems will not only reduce the performance of the battery and shorten the cycle life, but also may cause short circuits in a closed environment, causing safety hazards. Research shows that improving the stability performance of zinc anodes significantly improves the overall performance and safety of AZIBs.

This article reviews the limitations and challenges of zinc-negative poles in AZIBs, offering insights into future development prospects. These findings are aimed at guiding and promoting in-depth research on advanced nanomaterials of AZIBs.

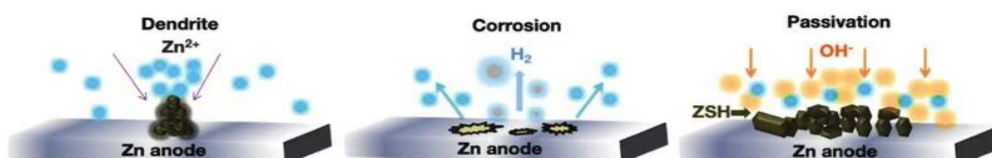
## 2. 2 RESEARCH ON ZINC ANODE ISSUES AND OPTIMIZATION STRATEGIES

**Dendrite growth:** Studies have shown that the main causes of dendrite growth are attributed to the uneven deposition of zinc and the instability of the electrochemical environment. Specifically, the surface of the zinc anode is not completely flat but has micro-scale concave-convex morphologies. These local protrusions or rough structures can cause uneven current distribution, leading to a significant increase in current density in specific areas. Due to the stronger electric field at the tips, local uneven electric fields are formed in these regions, which in turn promote the high enrichment of free  $\text{Zn}^{2+}$  at these tip positions, resulting in the preferential deposition of  $\text{Zn}^{2+}$ . This uneven deposition rate causes zinc to accumulate faster on the surface, thereby triggering the growth of dendrites [1]. The formation of dendrites may pierce the separator, causing a short circuit. Broken dendrites can form "dead zinc", reducing the charge-discharge performance of the battery [2].

**Hydrogen evolution reaction:** At the negative electrode of the battery, in addition to the zinc deposition reaction, various side reactions may occur, the most prominent of which is the hydrogen evolution reaction (HER) triggered by water molecules. As a typical electrochemical process, the kinetic behavior and reaction extent of this reaction are influenced by multiple factors, including electrode roughness, temperature, and electrolyte concentration [3].

The specific reaction process can be summarized as follows: Cathodic hydrogen evolution:  $2\text{H}_2\text{O} + 2\text{e}^- \leftrightarrow 2\text{OH}^- + \text{H}_2\uparrow$  [4].

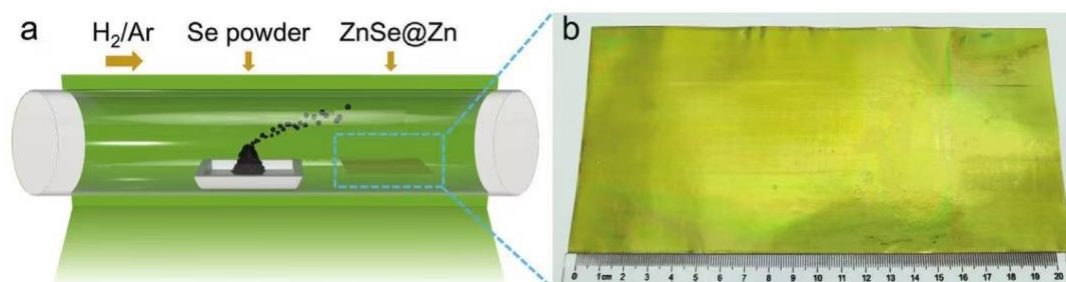
The hydrogen evolution reaction at the negative electrode occurs throughout the entire rest and operation period of the battery. The hydrogen evolution reaction consumes active zinc, reducing the coulombic efficiency. The hydrogen evolution reaction produces gas, which can cause the battery to expand or even leak. When the zinc negative electrode comes into contact with the electrolyte, many micro-scale galvanic cells are formed on a single zinc negative electrode, leading to the production of zinc hydrogen and fluctuations in the concentration of  $\text{OH}^-$  [5]. Corrosion occurs when locally concentrated hydroxide ions react with zinc ions to form a zinc hydroxide passivation layer, increasing the resistance of the interface and shortening the battery's service life [6].



**Figure 1:** Dendrite growth, corrosion, and passivation phenomena [7]

The above issues (as shown in Figure 1) are the main problems currently facing the development of zinc-ion battery anodes in aqueous systems. In response to dendrite growth, Corrosion, and passivation, researchers have proposed the following optimization strategies.

Interface modification layers play a crucial role in optimizing the negative electrode of aqueous zinc-ion batteries by precisely regulating the electrode interface. These layers primarily include inorganic coatings, organic-inorganic coatings, and nitrogen-doped carbon nanotubes, which work to stabilize the zinc negative electrode by regulating ion transport and deposition behavior.



**Figure 2:** Schematic diagram and macrograph of CVD growth of a uniform ZnSe layer on zinc foil [9]

**Inorganic coating:** Mainly inhibits dendrite growth by regulating zinc ion deposition behavior [8]. Inorganic coatings have special crystal structures or surface characteristics that can provide uniform nucleation sites for zinc ions, enabling Zn to form a more uniform and dense deposit layer during charging and discharging. Yang [9] et al. constructed a ZnSe nurturing layer on the surface of a zinc metal anode via in situ growth (as shown in Figure 2), providing a new approach for interface regulation of zinc anodes: the ZnSe nurturing layer possesses suitable crystal structure and chemical stability, enabling rapid and uniform transport pathways for  $\text{Zn}^{2+}$ , establishing stable ion transport channels, reducing nucleation sites for dendrite formation, and suppressing dendrite growth at its source. The in situ grown ZnSe layer exhibits excellent interface adhesion with the zinc foil substrate, improving the wettability of the electrolyte on the zinc anode surface. This allows the electrolyte to spread more uniformly across the electrode surface, ensuring an adequate supply of  $\text{Zn}^{2+}$  across the entire electrode surface and avoiding uneven deposition issues caused by poor electrolyte contact in localized areas, thereby further promoting uniform zinc deposition. ZnSe has stable chemical properties and is unlikely to react with components in the electrolyte (such as free water). It acts as a barrier, isolating the zinc foil from active substances in the electrolyte and significantly reducing the probability of side reactions, such as hydrogen evolution and zinc corrosion. Han et al. [6] utilized  $\text{NH}_4\text{F}$  as a precursor to construct a thin and uniform fluoride artificial interface (primarily composed of  $\text{ZnF}_2$ ) on the zinc metal anode surface, providing a key solution for enhancing the reversibility of the zinc anode and the performance of Zn/ $\text{MnO}_2$  batteries. This artificial  $\text{ZnF}_2$  interface possesses a unique crystal structure that enables rapid and uniform  $\text{Zn}^{2+}$  migration through an interstitial diffusion mechanism. Such an ordered diffusion mechanism prevents localized accumulation of  $\text{Zn}^{2+}$  on the electrode surface, thereby reducing dendrite nucleation and growth caused by uneven ion transport and ensuring homogeneous  $\text{Zn}^{2+}$  deposition from a kinetic perspective. Compared to unprotected zinc foil, this interface enables more stable  $\text{Zn}^{2+}$  diffusion rates and a more

E-controllable deposition process.

**Organic-inorganic hybrid layer:** This hybrid layer provides a uniform transmission channel for zinc ions and guides their uniform deposition. The organic component in the hybridization layer isolates the zinc negative electrode from the aqueous electrolyte, thus reducing the side reaction. Some functional groups in the hybridization layer can capture hydrogen ions and inhibit zinc cathode corrosion. Some hybridization layers can also regulate the solvation structure of zinc ions and promote the desolvation process, so as to achieve more uniform zinc ion deposition. Zhang et al. [10] designed a new type of hydrated eutectic electrolyte, using the dissolution reduction reaction between eutectic molecules and  $\text{SnI}_4$  to form an organic-inorganic hybridized SEI layer with gradient zinc affinity in situ on the surface of the zinc negative pole. Eutectic molecules play a key regulatory role in electrolytes, which can not only regulate the solvent environment of electrolytes but also affect the solvation structure of zinc ions.  $\text{SnI}_4$  undergoes dissolution-reduction reaction in the system, and its products participate in the construction of the SEI layer. During the charging and discharging cycle, these components interact to gradually build a unique SEI layer on the surface of the zinc negative pole. The gradient zinc affinity of the SEI layer is its most prominent feature: from the side closest to the surface of the zinc cathode to the side facing the electrolyte, the zinc affinity exhibits a systematic change. The area close to the zinc cathode has high zinc affinity, which can effectively guide zinc ion deposition and promote uniform nucleation growth; while the area close to the electrolyte has relatively low zinc affinity, which helps to prevent the excessive accumulation of zinc ions at the interface and inhibits the formation of branch crystals. Li et al. [11] constructed an organic-inorganic double-layer protective film (Nafion/ $\text{Zn}_3(\text{PO}_4)_2$ , NF-ZP) on the surface of the zinc electrode, which is composed of the upper organic layer (Nafion) and the lower inorganic layer ( $\text{Zn}_3(\text{PO}_4)_2$ ). This double interface layer can simultaneously inhibit the growth and side reactions of branch crystals. The inorganic  $\text{Zn}_3(\text{PO}_4)_2$  layer provides uniform nucleation sites, guiding the uniform deposition of zinc ions and inhibiting the formation of branch crystals; the organic Nafion layer blocks the direct contact between water molecules and zinc electrodes, minimizes side reactions, and still allows zinc ions to pass through to maintain the electrochemical properties of the battery.

**Nitrogen-doped carbon nanotubes:** A physical coating of nitrogen-doped carbon nanotubes (N-CNT) is used to protect the surface of zinc anodes. By adding nitrogen (N), more nucleation sites for zinc deposition are provided, which can regulate the electric field distribution on the anode surface, suppressing dendrite growth and the occurrence of side reactions. After comparing the phase composition analysis and electrochemical performance analysis of N-CNT, CNT, and BZ, the experimental results indicate that the N-CNT coating exhibits significant advantages on the zinc surface. N-CNT effectively suppresses the formation of dendrites on the zinc surface, which not only improves the quality of zinc deposition but also prevents the generation of byproducts during the charging and discharging process of the zinc battery [12].

The primary function of structural design is to optimize the microenvironment for zinc deposition in aqueous zinc-ion batteries.

**Bionic structure based on foam copper:** Kang et al. [13] and colleagues used chemical etching technology to build a three-dimensional porous copper frame on the surface of copper foil. This three-dimensional porous copper structure with rich pores and high specific surface area characteristics can effectively homogenize the electric field distribution and provide sufficient space for zinc deposition. The Hou team [14] reported that the composite zinc anode was prepared by molten penetration: the foam copper clamp was placed between two zinc foils, heated to  $450^\circ\text{C}$ , and then cooled to form a  $\text{PCu@Zn}$  anode. The process realizes the firm combination of zinc metals in the three-dimensional foam copper frame. The copper-zinc alloy layer formed can effectively guide zinc deposition, and the outer layer of zinc oxide protective film can prevent electrolyte corrosion. This multi-component synergy significantly improves the deposition/stripping stability of zinc anodes, so that the electrode can achieve a life of more than 1,000 cycles at a high current density of  $10\text{mA cm}^{-2}$  [15].

The main purpose of electrolyte regulation is to optimize the structure and chemical interface of the solvent in aqueous zinc-ion batteries.

**Electrolyte regulation:** The basic principle of electrolyte regulation engineering in water-based zinc-ion batteries is to solve key problems such as zinc branch crystal growth, dehydrogenation side reaction, and insufficient electrolyte stability by optimizing the physicochemical properties, interface reaction, and ion transmission behavior of electrolytes, and finally improve the cycle life, Coulomb efficiency and safety of the battery. Wang [16] discovered that N-hydroxy phthalimide can serve as a functional electrolyte additive to form a hydrophilic-hydrophobic interface layer (HHIL) in situ on the surface of the zinc negative electrode through  $\pi$ - $\pi$

interaction and ion-dipole interaction. The interface layer promotes the formation of an inorganic inner layer rich in ZnS and ZnF<sub>2</sub>, and realizes the synergistic effect of the flexible molecular self-assembly layer and the rigid inorganic inner layer. While effectively inhibiting side reactions and branch crystal growth, it significantly improves the controllability of Zn<sup>2+</sup> deposition. The Zn//Zn symmetrical battery built based on this design has achieved a stable cycle of more than 900 hours under high current density conditions (20 mA/cm<sup>2</sup>, 10 mAh/cm<sup>2</sup>) [17].

### 3. FUTURE PROSPECTS OF AQUEOUS ZINC-ION BATTERIES

This article summarizes the challenges faced in the development of aqueous zinc-ion batteries and the recent research on anode improvement strategies. Although certain progress has been made in addressing issues such as anode dendrite growth and side reactions in current aqueous zinc-ion batteries, they still encounter challenges like energy density bottlenecks and insufficient long-term cycling stability.

From a developmental perspective, future research could focus on multifunctional interfacial regulation techniques that combine the advantages of inorganic-organic hybrid layers to optimize ion transport and interfacial stability precisely. Additionally, exploring novel zinc-based anode materials, such as alloying and nanostructured designs, could enhance zinc storage capacity and structural durability. Integrating AI-assisted methods could accelerate the efficient development of electrolyte formulations and electrode architectures, driving aqueous zinc-ion batteries toward higher energy density, longer lifespan, and lower cost. These advancements are expected to enable broader applications in large-scale energy storage and portable electronics, providing critical support for the construction of new energy systems.

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